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Final Report

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Optical/Far-Infrared Control of Low-Dimensional Semiconductor Structures*

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$500,000

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*This program was transferred to Georgia Tech from Washington State University where it was previously entitled "Coherent Control of Carriers and Light in Semiconductor Nanostructures."

Introduction:

A program of research was carried out in which theoretical investigations into the simultaneous manipulation of carriers (electrons, holes, and ultimately excitons) and light in semiconductor nanostructures such as quantum wells were conducted. The manipulation of the carrier and optical dynamics will be achieved by the use of specially tailored ultrafast optical pulses, multicolor laser fields, millimeter or submillimeter electromagnetic pulses, or combinations of the above. Because the relevant timescale for the carrier dynamics may be less than the characteristic dephasing time of the carriers, the evolution of the system can be coherent; phase effects play a dominant role. Such shaped pulses and multicolor fields may be used to coherently control optical excitations in semiconductors in order to access quantum mechanical states, which are otherwise difficult to attain.

The project fell under the ONR Young Investigator Program (YIP) and the Presidential Early Career Award for Scientists and Engineers (PECASE).

Work for this project began at Washington State University under the title "Coherent Control of Carriers and Light in Semiconductor Nanostructures," and was subsequently transferred to Georgia Tech as "Optical/Far-Infrared Control of Low-Dimensional Semiconductor Structures."

Research Highlights:

I. Low Carrier Densities in Intrinsic Semiconductor Heterostructures: Strong-Field Terahertz Physics of Excitons in Low-Dimensional Semiconductor Structures: We studied theoretically the transient response in the THz and optical domains of an intrinsic semiconductor quantum well (QW) *weakly* excited by an ultrafast optical pulse whose spectral bandwidth spans excitonic levels as well as free e-h pairs in the presence of a *strong* THz field. Our studies began with the field polarized in the QW plane, in which case the THz field coupled different states of the internal exciton motion *[Citrin(c),(f),Hughes(a)]*. These dynamically hybridized states are then interrogated by a weak optical (near-infrared) field. The initial focus of our work was the coherent control of the emitted THz transients *[Hughes(e),(f)]*. THz harmonic generation was predicted there—similar to high-field harmonic generation (HFHG) in atoms *[Corkum,Kulander]*. We also studied the optical properties, such as the appearance of THz sidebands on optical spectra *[Kono]*. These are optical signals at frequencies $\omega + n\Omega$ where ω is the incident optical frequency, Ω is the THz frequency, and n is an integer. We have investigated the THz fields at which perturbation theory breaks down: the kV/cm range at ~1 THz in good agreement with experiment *[Sherwin]*. Also considered was an in-plane circularly polarized THz field; we have found that the sidebands are strongly suppressed for the circularly polarized case, in excellent agreement with experiment *[Kono]*.

It was found in Prof. Mark Sherwin's group at UCSB, however, that more efficient THz-sideband generation takes place if the THz field is polarized in the QW growth direction. Our estimates for the maximum frequency conversion to the first THz sideband normalized to the peak reflection at the fundamental (optical) frequency is a few % in a single QW *[Maslov(a)-(c),(e)-(h)]*. Such conversion efficiencies thus make THz-sideband generation in QW's potentially attractive for optical wavelength conversion or to shift an optical data stream in a WDM system from one wavelength to another. This work proceeded in collaboration with Prof. Mark Sherwin at UCSB, and continues.

To understand basic issues involved in THz-sideband generation, we have developed and implemented simple theoretical models. Full-scale calculations of the optical properties of

QW's in THz fields including the detailed e-h Coulomb interaction are computationally expensive, and, moreover, can obscure the important physical effects in oceans of numbers. Instead, analytical approaches are useful to explore parameter space as well as to identify effects observed in more detailed simulations *[Citrin(n)]*. The treatment is equivalent to the standard approach to single-particle quantum transport through a time-modulated potential *[Citrin(l),Glazman,Rubo,vanHouten]*. The result is the analogy between the single-particle transport and optical phenomena *[Citrin(k),(m)]*.

Initial numerical results for a single QW show that the first sideband is in the range of 1 % the peak height of the fundamental; this is in excellent agreement with detailed computations we have carried out. [Subsequent work (as discussed above) established theoretical maximum conversion efficiencies in the few % range.] For the time-domain problem—using ultrafast optical pulses to photogenerate excitons—we have shown how a quasi-half-cycle THz pulse can be used to manipulate the optical phase of the excitons, and thus how to carry out in principle two-pulse coherent control of excitons in a QW *[Citrin(a),(b),(d),Heberle,Luo]*.

The treatment has been generalized *[Citrin(l)]* to account for confinement of the optical field in a planar semiconductor MC *[Weisbuch]*, where a bare-cavity optical mode is degenerate with an excitonic resonance. The resulting two coupled modes of the MC are of hybrid exciton—cavity-photon character; mode splittings of ~1-10 meV are typical. We have shown that THz-sideband generation can be strongly enhanced by two orders of magnitude in a MC *[Citrin(l)]*. We have also considered the corresponding problem in the time domain *[Norris]*, where an ultrafast optical pulse excites the MC which is followed by an ultrafast THz pulse. We have shown how such a THz pulse can be used to control the phase of the oscillations between the two modes excited simultaneously by the ultrafast optical pulse.

Another model explored is a pair of excitonic resonances dressed and resonantly coupled to each other by the THz field *[Citrin(i)]*. The coupling results in the spectral features associated with single unmodulated resonances becoming doublets as the modulating field is applied [Autler-Townes (AT) splitting]. At sufficiently high THz fields, a hole at the center of the doublet forms leading to electromagnetically induced transparency (EIT). So far as we know, we are the first to study the AT splitting and EIT in the THz sidebands *[Citrin(i)]*.

Using many of the techniques we developed for the foregoing topics, we also considered the optical propeoreties of QW's in strong magnetic fields or strong crossed magnetic and THz electric fields in the nonperturbative regime *[Citrin(h),Hughes(i)]*. The theory well reproduces detailed theoretical spectra in limits where they can be obtained, such as for the in-plane dc *[Franz,Keldysh]* and dynamic Franz-Keldysh effects *[Nordstrom(a),(b)]* (the modification of the bandedge optical properties in a dc or THz electric field) and magnetoexcitons *[Glutsch]*.

We conducted work on the optical properties of quantum wires (QWR's) in strong dc electric fields aligned with the structure axis *[Hughes(d)]* using a real-space approach. We found that the Franz-Keldysh effect including excitonic effects in QWR's can be quite dramatic *[Hughes(d)]*.

<u>II. High Carrier Densities in Semiconductor Optical Amplfiers: Strong-Field Terahertz Effects on the Optical Properties:</u> We studied the transient dynamical response of semiconductor optical amplifiers (SOA's) to half-cycle *[Hughes(b),(c)]*and narrow-band *[Ning]*THz pulses. We have found a substantial THz-induced heating effect of the e-h plasma in the SOA with cooling via LO-phonon emission to the lattice temperature within ~5 ps. The heating-induced modification of the gain spectrum and frequency-dependent refractive index are predicted to lead to substantial modification of the propagation of an optical pulse through the SOA over lengths of ~300 μm for achievable THz pulses, thus suggesting interest for possible applications ultrahigh-speed optical switching. Initial experiments carried out by Prof. James

Heyman of MacCalester College, MN conducted at the UCSB were inconclusive, and follow-up experiemtns are planned.

II. Coherent Control of Excitons in Quantum Structures: *Coherent control* is the use of multicolor electromagnetic fields, or shaped optical pulses, to achieve quantum states that may be otherwise difficult to attain. One application of these ideas in semiconductors is to use phase-locked pairs of optical pulses, first to excite and then to deexcite excitons. If the two pulses are chosen with a time delay such that their phase difference is an odd multiple of π, the two pulses excite interband polarizations that are π out of phase, and thus cancel. This method has been applied to QW's on the ps timescale *[Brener,Heberle,Luo,Planken]*, which is much shorter than the exciton radiative lifetime. We have recently proposed an ultrahigh-speed semiconductor-MC-based switch based on this principle to circumvent the difficulties associated with slow device recovery (saturation) *[Citrin(a),(b),(d)]*. Thus the recovery time as well as the switching time is expected to be consistent with 100-Gb/s applications. Together with our experimental collaborators, we have recently demonstrated the physical principle for such a switch *[Lee]*. We have treated the linear and nonlinear interactions of the optical pulse with the QW's *[Lee]*. In particular, we have studied theoretically the optical nonlinearity viewed at one coupled mode of the microcavity after pumping at the other mode using a phase-locked pulse pair. Substantial modulation of the probe reflectivity was found in agreement with experiments carried out in collaboration with Prof. Ted Norris at the University of Michigan. These calculations are based on the time-space domain solution of Maxwell's equations in the presence of the nonlinear medium employing the finite-difference time-domain method *[Sullivan(a),Taflove]*. The material equations are the optical Bloch equations with phenomenological nonlinearities.

IV. Electronic Wavepackets in Superlattices: The dynamics of electronic wavepackets in SL's driven by strong time-dependent THz fields was investigated. Some of the phenomena of interest were Bloch oscillations *[Bloch]*, collapse of the miniband *[Holthaus]*, and Zener tunneling *[Zener]*. In particular, we considered the generation of multiple harmonics by electrons in a superlattice in a Kronig-Penney model numerically by means of FDTD, and analytically by semiclassical transport theory *[Feise(a)]*. We found the cutoff order of the emitted THz harmonics is simply the ratio of the potential energy drop at THz field maximum per supercell of the superlattice to the energy per THz photon—quite distinct in nature from HFHG in atoms. This cutoff is simply determined by the maximum kinetic energy an electron in a miniband can acquire from the THz field before it undergoes Bragg scattering (Bloch oscillation).

V. Carrier Dynamics in Photoconductors: Our interest is in the interplay of the carrier dynamics, electrode and excitation geometry, and screening in fast photoconductors excited by ultrafast optical pulses to optimize THz output from such devices. We have carried out work with our experimental collaborator Prof. Martin Koch of the Technical University of Braunschweig, Germany to study the spatio-dynamics of optically excited carriers in photoconductors. Semi-quantitative agreement has been achieved. We have found that the space charges persist under 80 MHz repetition-rate operation, which is standard for photoconductors pumped by Ti:Sapphire oscillator systems *[Bieler,Feise(b)]*. Currently, a graduate student is continuing work on this topic.

VI. Miscellaneous: We have carried out a number of studies related to the foregoing but not falling neatly into any of the categories. One is excitonic Rabi flopping in QW's *[Geissen, Schultzgen]*, in which a strong optical pulse coherently drives the exciton population first up and then back down. We have studied carrier dynamics upon the application of strong sub-ps optical pulses using the SBE in the HF approximation with non-diagonal scattering. In good agreement with experiment, we find deep modulation of the carrier population. We have proposed carrying Rabi-flopping experiments in a dc biased QW to produce strong, tunable THz emission which follows the Rabi flops *[Hughes(j),(m)]*. We have also studied quasi-adiabatic population transfer using strongly chirped or frequency-modulated optical pulses incident on QW's. We have found conditions under which the carrier populations are transiently pinned at a roughly fixed value while the pulse is incident, as opposed to inducing Rabi flops *[Hughes(l)]*. Also partially funded by this grant was work on Rabi flopping in two-level systems induced by few-cycle optical pulses where it was found that for sufficiently strong pulses the area theorem breaks down *[Hughes(k)]*.

Other work under the ONR program includes electron-phonon interactions in QD's *[Goupalov]* in a collaboration with Profs. Hailin Wang at the University of Oregon, Robert Suris at the Joffe Institute, St. Petersburg, Russia, and P. Lavallard at CNRS, France. We have also performed model calculations for THz-modulated QWs *[Citrin(r)]*, of THz nonlinearities *[Citrin(t)]*, electron-hole spatial correlation in microcavities *[Citrin(s)]*, and on a formalism that might show promise for semiconductor nonlinear-optics problems *[Setlur]*.

Another area which have attracted our interest are the electromagnetics of time-varying plasmas in semiconductors for electromagnetic frequency conversion *[Bakunov(a)-(c)]* and fundamental issues concerning the relaxation of carrier distributions that are anisotropic in *k*-space, such as are generated by THz pulses *[Hughes(n)]*.

We have also worked on nonlinear dynamics of electrons in QW's in THz fields *[Batista(a), (b),Citrin(q)]*, nonlinear optics of plasmas in solids *[Bakunov(d)]*, THz pulse shaping *[Nekkanti]*, and quantum dynamics in QD's *[Sullivan(b),(c)]*.

Personnel:

Funding from this program supported at various times two Ph.D. students (Alexey Maslov, PhD 2001, subsequently post-doc in PI's group at Georgia Tech, now post-doc at NASA Ames, Moffett Field, CA; Michael Feise, PhD 2001, subsequently post-doc at Washington State University, now post-doc at Australia National University) and partially supported four post-doctoral research fellows (Stephen Hughes, subsequently lecturer at University of Surrey, then research staff at Galian Photonics, Vancouver, BC, now member research staff, NTT Research Laboratories, Tokyo; Alex Maslov, see above; Adriano Batista, now competing for faculty position in Brazil; Girish Setlur, post-doc at Indiian Institute of Science, Bangalore, India; Sergei Goupalov, now post-doc at Los Alamos National Laboratory). The program also provided partial summer support for the PI.

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Intellectual Property:

No patents, licenses, or disclosures of invention resulted from this program.

Subcontracts:

None.